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Attempts to Produce D2-Gas-Filled Be Shells

Bob Cook, Mike McElfresh, Craig Alford, Eric Fought, Steve Letts

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From: Bob Cook, Mike McElfresh, Craig Alford, Eric Fought, and Steve Letts

Subject: Attempts to Produce D_2-Gas-Filled Be Shells

Summary

We have attempted to fabricate some 0.5 mm diameter D_2-gas-filled Be shells by coating gas-filled PVA-coated GDP mandrels with Cu-doped Be. We find that during the coating all (or most) of the gas leaks out. This is likely due to either small cracks or holes in the coating that are formed at the earliest points and are maintained during the thickness build-up of the coating, and / or to some level of intrinsic porosity in the coating. This memo documents our efforts.

Introduction

Given the emergence of the graded Cu-doped Be capsule as the lead candidate for ignition experiments in 2009-2010, there is interest now in gaining experience with Be as an ablator in convergent geometries. Cu-doped Be can be sputtered onto plastic mandrels, and composition gradients can be formed if desired.[1] The primary current problem is in filling capsules with gas. Plastic capsules have historically been filled by diffusion, Be capsules cannot be. We have demonstrated that we can drill very small fill holes in Be capsules, and thermally remove the mandrel,[2] but at this moment we have no way of sealing the capsules after fill. Work is in progress at General Atomics to develop a method of laser sealing the holes in gas-filled shells by melting a small region of the capsule around the hole.[3]

An alternative approach that we have tried is to coat already filled plastic mandrels. We need to emphasize that this approach, which will be detailed here, had several drawbacks before we started. First, such capsules would have a plastic inner shell, at least 6 μm in thickness. Second, the fill attainable would be limited by a combination of the limited strength of the initial mandrels and their leak rate at the temperature necessary to put a sealing layer of Be on them. Third, we would have no way to measure the actual fill of the capsules which would be shot, the best we would be able to do is get an estimate by destructively measuring the fill of their siblings. However the reality is that all of these drawbacks were of no concern since we found

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[1] J. Gunther, M. McElfresh, C. Alford, H. Huang, and B. Cook, "Fabrication and Characterization of Graded Cu-doped Be Shells - Details and Documentation of Our First Attempt," LLNL technical memo, October 12, 2004. Copy available from Bob Cook.

[2] B. Cook, S. Letts, S. Buckley, "Experimental Confirmation of CH Mandrel Removal from Be Shells," LLNL technical memo, June 8, 2004. Copy available from Bob Cook.

[3] N. Alexander, General Atomics.

experimentally that we could not coat the filled capsules and form a Be seal that prevented the fill from leaking out. What follows is a fairly chronological version of what was attempted, and the conclusions that we draw from this failed effort.

Results

As noted above, the basic approach was to sputter coat Be onto filled mandrels. Since simple GDP has a diffusion half-life measured in minutes, we needed to work with PVA coated mandrels which at room temperature have half-lives of 20 or more hours. Thus the initial questions were 1) what fill pressure might we be able to get without bursting the shell and 2) what would be the leak rate under the coating conditions where elevated temperature is a factor. The experience at GA was that 3 μm thick 1 mm diameter shells have a burst pressure of about 10 atm, thus at 0.5 mm diameter we might expect a burst pressure of 20 atm. Since the GDP mandrels would have up to 3 μm of PVA on them as well this should provide extra strength. However the 20 atm limit is at room temperature, at elevated temperature the internal pressure will be higher, as well as the material weaker. Lastly it is well known that the gas fill half-life due to PVA decreases markedly with increased temperature. Though it has never been measured directly for D_2, it has for Ar, and the half-life is about a day at 110 °C and about an hour at 142 °C.[4] If D_2 has the same temperature dependence, i.e. the half-life dropping by a factor of 24 every 32 °C, then a shell which had a half-life of 1 day at 25 °C would have a half-life of 1 h at 57 °C or a couple of minutes at 89 °C. Clearly we want to keep the initial temperature that the filled PVA coated shells see as low as possible while coating them with Be as fast as possible. Unfortunately fast Be coating means higher temperature, thus a balancing needed to be achieved. Since we initially thought that even a very thin (few nm?) coating of Be should stop permeation, we chose to try initially to keep the temperature as low as possible.

In order to test our approach we got some PVA coated 0.5 mm GDP mandrels from GA filled with 0.5 atm of Ar. These shells also had about a μm of GDP over the PVA to protect it. Thus the total wall thickness was about 7 μm (3 μm GDP, 3 μm PVA, and 1 μm GDP). Our thought was that if all the Ar (which can easily be measured by XRF) is lost during the coating then certainly the D_2 will also be lost. Since we initially were only interested in gas retention opposed to strength we had these shells filled with 10 atm of D_2. The shells were coated with Be for 6 h at a low power setting, resulting in about 0.2 μm of coating. We believed this should have been enough to seal the capsule. XRF measurement showed that the Ar was still in the capsules and that the level had not changed. Four capsules were sent to GA for bursting in oil to measure the fill by the dimensions of the bubble that was formed. The initial results from this characterization were mixed, 2 shells were broken (6 days after removal from the coater) and were found to contain 3.2 and 2.8 atm of gas. Dave Steinman at GA had the presence of mind to hold back the other two shells for later breakage to confirm that the fill was not leaking through the Be coating. However the capsules continued to leak because when he broke them at 12 days the yield was again reduced. The data is shown in Figure 1. If one fits the data to

[4] D. Steinman, General Atomics.

$$P(t) = P(0) \cdot \exp(t / t_{1/2}) \quad (1)$$

one finds that the fill at $t = 0$, i.e. the time the shells came out of the coater, was about 7.5 atm and that the half-life was about 6.5 days.

So why were they leaking? We believed that diffusion through contiguous Be metal was probably not happening. Several years ago we explored the diffusion of hydrogen through Be shells and concluded it to be non-existent even at elevated temperatures. We reasoned that probably the layer was so thin (~0.2 μm) it may have developed cracks due to pressure variations across the capsule wall from the temperature decrease due to the cessation of coating coupled with the increase in external pressure when removed from the coater. The positive result was that the shells probably had a 7.5 atm fill upon being removed from the coater, the loss from 10.5 atm (10 atm of D_2 fill and 0.5 atm of Ar) due to the temperature exposure in the coater.

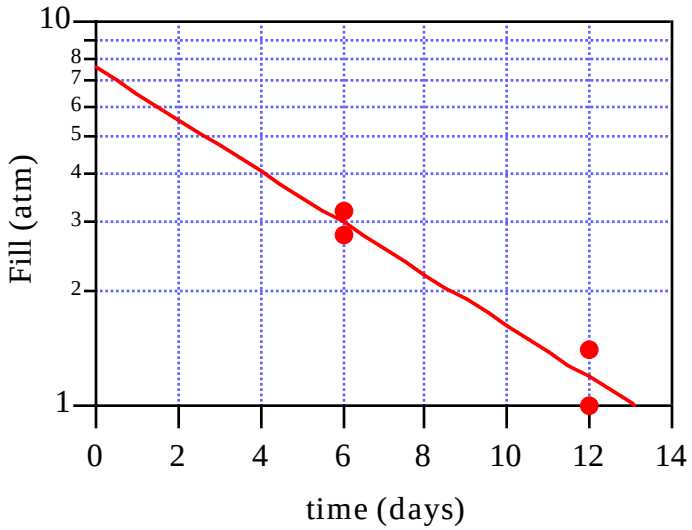

Figure 1. Fill as a function of time in first experimental shells with a ~0.2 μm coating of Be.

In order to check this out a second experiment was done and a much thicker coating of Be applied before the shells were vented to atmosphere. For this experiment about 2 μm of Be was applied to the filled (10 atm D_2, 0.5 atm Ar) shells. Four shells were sent to GA for testing. Two shells were broken immediately and had fills of 8.1 and 9.0 atm. The other two were held at 35 °C for 9 days before bursting to magnify any leakage, when broken they gave 7.3 and 7.6 atm. The relatively small decrease might have been due to a small amount of leakage due to cracks, or it might simply have been a result of limited statistics. In any case we felt confident that the coating process could be performed on PVA-coated gas-filled shells with substantial retention of the gas fill.

In order to assess a coating procedure as quickly as possible, as well as making it possible to evaluate the retained fill at intermediate steps of coating, we decided to build a system at LLNL for checking the gas pressure remaining in the capsules. The system shown schematically in Figure 2 is similar to one used at GA. The vacuum pump we

used was part of a system that included a residual gas analyzer that allows us to identify and distinguish between hydrogen, argon, and various possible contaminants (e.g. water and air).

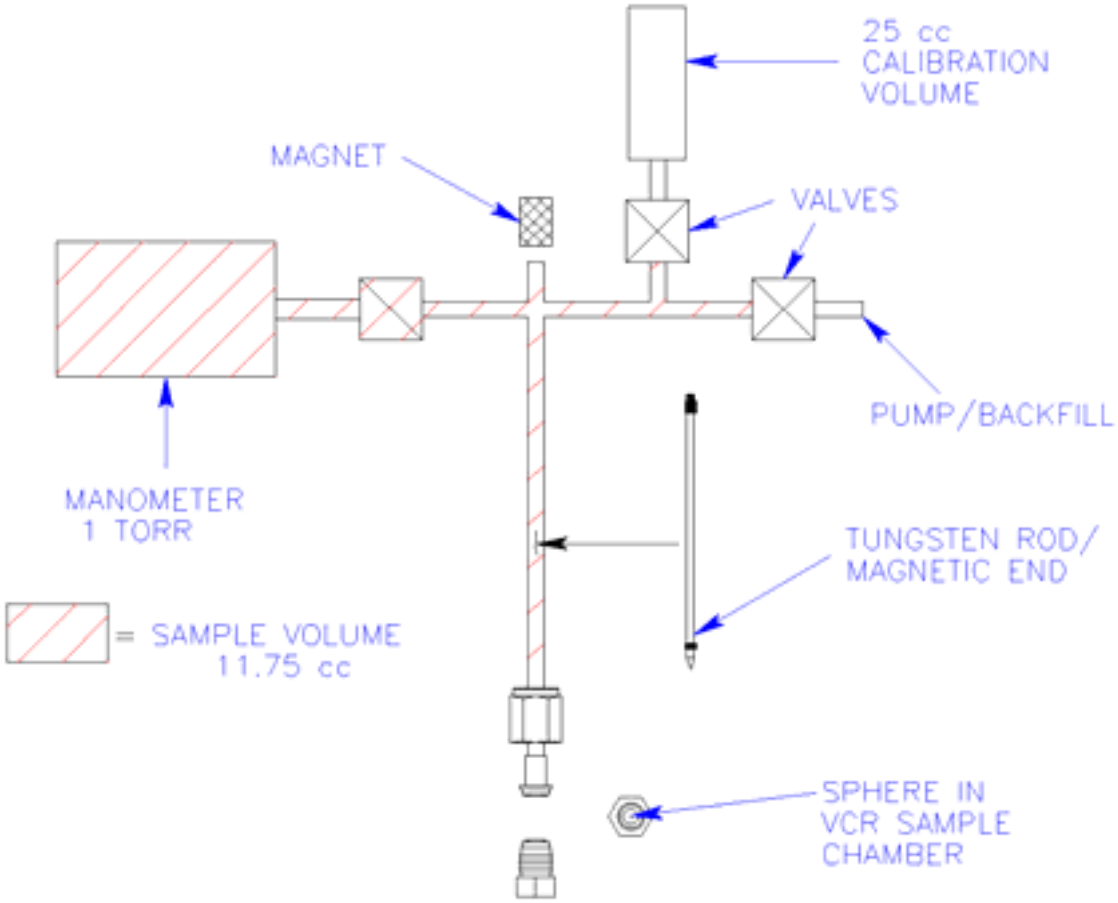

Figure 2. The system shown above is used to measure the gas pressure inside small spheres. A sphere is placed in a counter bore machined in the male VCR fitting. The system is evacuated. The magnet is then removed allowing the Tungsten rod to side down the inside of the tube and break the sphere. The pressure is measured with a one Torr capacitance manometer.

We next proceeded to the mandrels that had been prepared by GA for the Omega experiment. These were filled to 15 atm with D_2. The general coating steps were 1) a 2 to 6 hour precoat using the dimpled precoat pan with two Be guns operating at 40 to 50 watts each, 2) move the capsules to the regular pan and coat using four Be guns at 100 watts each for 6 hours, 3) ramping the power to a higher level over several tens of minutes to the full desired thickness. Initially the sputter guns were shut off over night, however in later runs the guns were run continuously 24 hours per day. After the first coating run showed that little gas was retained, we worked under the assumption that reducing the temperature of the capsules during deposition would be the best way to maximize the chance of retaining a significant fill.

15-atm Runs

The initial run involved loading four 0.5 mm capsules each with a 15-atm fill and pumping overnight, then the next morning a precoat in the dimpled pan for 6 hours with 2 guns at 50 watts each was run. The system was shutdown overnight, the chamber opened the next morning and capsules moved to a smooth pan, pumped down for 2 hours and then the power on 4 guns was ramped over 45 minutes to 100 watts and the capsules coated for 6 hours. The system was then shutdown overnight with chamber pumped and the next morning the power on 4 guns was ramped over 90 minutes to 300 watts each and then run for 2 hours. The system was allowed to cool, capsules removed and tested. The capsules were found to be empty.

The second run was different only by eliminating the 2-hour coating at 300 watts per gun. Instead the last step involved reaching 300 watts per gun but shutting off the system immediately after reaching 300 watts. Final pressure values on the capsules of 1.8, 2.0, 3.0, and 5.4 atm were determined.

After these failures an attempt to get as much Be on the capsule as quickly as possible was attempted. So the Be gun power was ramped to 300 watts (4 guns) over 5 minutes. The capsules exploded after 6 minutes at 300 watts.

The next attempt involved eliminating the overnight shutdown in order to avoid the temperature excursions associated with turning off the guns. We have evidence that the capsules expand during deposition and this thermal cycling may be compromising the integrity of a thin Be shell. So Be gun power was set to 50 watts with 2 guns for 4 hours, then without stopping the power was ramped over 45 minutes to 100 watts and then held for 4 hours, and then ramped over 45 minutes to 200 watts and held for 4 hours. This run demonstrated the possibility of running 24/7. A capsule was tested before deposition and shown by our measurement method to hold 14.6 atm. At the end of the run a capsule was shown to have 1.5 atm remaining.

Being unable to retain gas in the coated capsules, and assuming that the additional pressure associated with capsule heating may be the culprit, we decided to attempt to monitor the temperature of the capsules during deposition. A thermocouple was positioned just over the bounce pan. The plot in Figure 3 shows sputter gun power and the associated temperature as a function of time. Initially two sputter guns operating at 50 watts, as is the case during precoating, were turned on. After 20 minutes all four guns were operated at 50 watts, then the power on all four guns was stepped up in 50-watt increments every 20 minutes. There are several curious aspects to these results. First there appears to be a rapid increase in temperature when the power is turned up to 150 watts. But even more curious is the leveling off, and even decrease, in temperature at the highest power settings. Additionally, it should be noted that it is difficult to know the actual temperature of the capsule since it is making intermittent contact with the pan while the thermocouple has a constant link with a thermal sink.

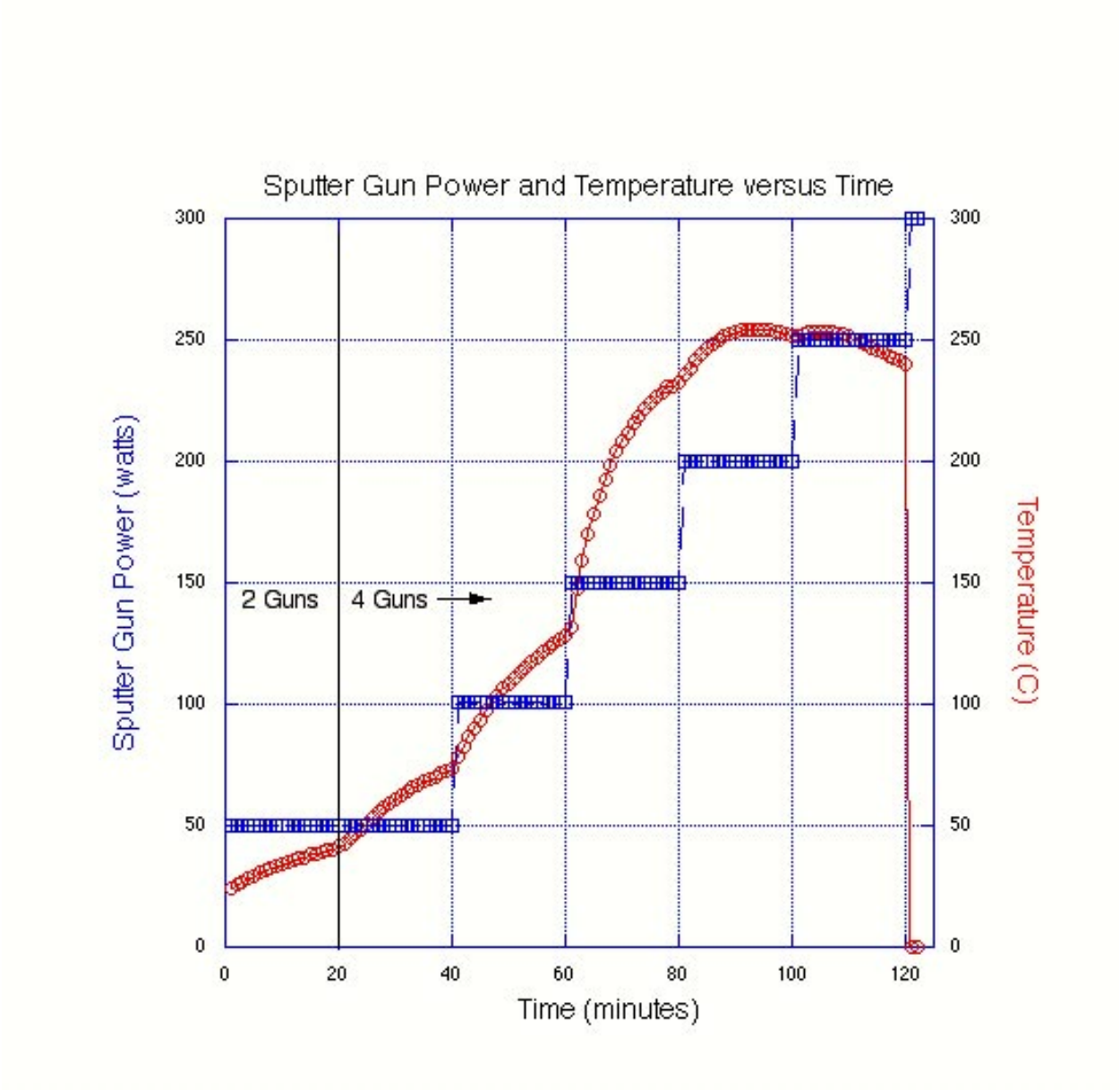

Figure 3. Using a thermocouple placed just above the bounce pan, the temperature was measured at different values of sputter gun power and at 50 watts for both two and four guns operating.

The next run we returned to a procedure similar to the initial run (i.e. precoat on dimpled pan and transfer to smooth pan) except that the 300-watt run was eliminated and the 100 watts per gun with 4 guns was run for 42 hours (in order to stay below the large temperature increase at 150 watts). Before coating one capsule was broken and found to have 11.7 atm of fill. At the end of the run the capsules were shown to have less than 1 atm of fill. The RGS showed the gas to be primarily argon.

The final attempt for a 15-atm fill was similar to the previous run except that the capsules were held for 3 hours at 100 watts on four guns. A capsule was removed and burst to find 3.5 atm of fill.

After these failures to retain gas capsules filled with 15 atm, the fill pressure was returned to 10 atm as had been used in the earliest runs. As a final attempt the power was held at 50 watts. A precoat was done in a smooth pan (rather than the dimpled pan) with 2 guns at 50 watts each for 6 hours. The next day then the power on 4 guns at 50 watts was run for 69 hours. The capsules were shown to have less than 1 atm of fill and the gas was argon.

Conclusions

The simple conclusion is that using our approach to we have not been able to seal in D_2 using a sputter-deposited Be layer. The reasons for this are not entirely clear, however. Clearly the D_2 continues to "leak" through the Be coating after a thin coating is applied. Whether this is due to an actual porosity in the sputtered Be, or due to the formation of cracks due to a combination of the fill and the temperature excursions the shell experiences, or both, is not clear. However the porosity of at least thin layers of Be is supported by some unexpected results from another experiment. As noted earlier, we have developed a method of removing the plastic mandrel by pyrolyzing it at elevated temperatures with air that is pumped in and out of a small, laser-drilled hole in the shell. As part of the development of this technique some roughly 10 µm walled shells were placed in a furnace at 450 °C with NO hole drilled. In the absence of O_2 the plastic mandrel will carbonize, forming H_2 and low hydrocarbon gases, along with solid carbon. Because the shell had no hole in it for these gases to escape, one would expect the mass of the shell to remain essentially unchanged. However this was NOT the case with these thin walled capsules. There was a significant mass decrease indicating that gasses were escaping through the shell wall. It is possible that the thin wall cracked at elevated temperature, and this allowed the gas to escape, but there was no obvious visual evidence of this. The same experiment (no hole) was tried with 20 µm walled shells and there was no mass decrease, indicating that at least at this thickness the wall is impervious to gasses.

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